Polonium-210 and Lead-210 in the Terrestrial environment: A historical review.

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Natural origin of ^{210}Po and ^{210}Pb in the terrestrial environment

Polonium-210 occurs widely in the terrestrial environment and as □-emitter and is an important component of man's natural radiation background. It's presence in deep soils and minerals may be traced to the decay of radionuclides of the ^{238}U decay chain:

$$^{238}\mathrm{U} {}^{>234}\mathrm{Th}>{}^{234}\mathrm{Pa}>{}^{234}\mathrm{U}>{}^{230}\mathrm{Th}>{}^{226}\mathrm{Ra}>\mathbf{{}^{222}Rn>}$$

Radon-222 originates from the decay of uranium-238 in the earth´s crust and diffuses from soil to the atmosphere where it´s concentration decreases monotony by height. With a half-life of 3.82 d **^{222}Rn** decays to the short lived radon daughters:

$$>{}^{218}\mathrm{Po}\ (\mathrm{RaA}\ 3.10\ \mathrm{min})>{}^{214}\mathrm{Pb}\ (\mathrm{RaB}\ 26.8\ \mathrm{min})\ >{}^{214}\mathrm{Bi}\ (\mathrm{RaC}\ 19.9\ \mathrm{min})>{}^{214}\mathrm{Po}(\mathrm{RaC}'164.3\ \mu\mathrm{s}).$$

These decay products attach to airborne particles which deposit as dry and wet deposition on the earth´s surface. ^{218}Po (RaA 3.10 min) is in radioactive equilibrium with **^{222}Rn** at about 5 m above surface and ^{214}Pb(RaB 26.8 min) >^{214}Bi (RaC 19.9 min)>^{214}Po(RaC´164.3 μs) is in radioactive equilibrium with **^{222}Rn** at about 50 m.

The concentration of the long lived decay products;

$$^{214}\mathrm{Po}>\ \mathbf{{}^{210}Pb}\ (\ \mathrm{RaD}\ 22.20\ \mathrm{a})>{}^{210}\mathrm{Bi}\ (\mathrm{RaE}\ 5.01\ \mathrm{d})\ >\ \mathbf{{}^{210}Po}(\mathrm{RaF}\ 138.4\ \mathrm{d})\ >{}^{206}\mathrm{Po}(\mathrm{stable})$$

increase with height and reach a maximum in the stratosphere. The activity concentration of ^{222}Rn and its short lived decay products at the earth´s surface (1-10m) is about 7-2 Bq/m^3 (W. Jacobi, 1963).

By simultaneous measurements of ^{222}Rn flux and gamma ray dose rate (GDR) at 63 locations in Switzerland, Germany, Finland and Hungary it was found that a relatively stable fraction (20%) of the total terrestrial GDR originates from the ^{238}U decay series, of which ^{222}Rn is a member. Accordingly, spatial variation in terrestrial GDR was found to describe almost 60% of the spatial variation in ^{222}Rn flux. Furthermore, temporal variation in GDR and ^{222}Rn was found to be correlated. Increasing soil moisture reduces gas diffusivity and the rate of ^{222}Rn

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ED_001091_00027922-00001

flux but it also decreases GDR through increased shielding of photons. Prediction of ^{222}Rn flux through GDR for individual measurement points is imprecise but un-biased. Verification of larger scale prediction showed that estimates of mean ^{222}Rn fluxes were not significantly different from the measured mean values.

The regression equation (Eq. (1)) reads:

$$\mathbf{^{222}Rn\text{-}flux[atom.cm^{-2}.s^{-1}] = a \,\square\square\text{-}dose\ rate\ [\square\ Sv\ h^{-1}]\ -\ b}$$

where a = 11.8 (SE ± 1.3); b = 0.15 (SE ± 0.11).
(T. Szegvary et al., 2009; T. Szegvary et al., 2007a; T. Szegvary et al., 2007b)

The activity ratio of $^{210}Pb/^{222}Rn$ in ground air is in the order of 10^{-4} and the $^{210}Po/^{210}Pb$ ratio in the order of 0.05-0.2 (M. Baskaran and G. E. Shaw, 2001; D. McNeary and M. Baskaran, 2007).

By studying the relation between the ^{222}Rn-flux and the $^{210}Po/^{210}Pb$ concentration in air and in fallout more carefully it might be possible to develop a model by which the $^{210}Po/^{210}Pb$ concentration in air and in fallout could be roughly estimated from □-dose rate measurements. Such a model could be verifies by simultaneous measurements of ^{222}Rn flux and ^{210}Po in air filters at the GDR locations in Europe (W. Jacobi, 1963).

Anthropogenic sources, such as coal combustion and nuclear explosions, have been computed to contribute less than 1% for ^{222}Rn and ^{210}Pb in the atmosphere (UNSCEAR, 1988).

Atmospheric fallout of ^{210}Po and ^{210}Pb

Since late 1950th observations of $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ activity ratios have been widely used to determine the mean residence time of natural aerosols in the troposphere (G. Lambert and M. Nezami, 1965; G. T. Piliposian and P. G. Appleby, 2003; C. Papastefanou, 2006; N. Rastogi and M. M. Sarin, 2008).

The results of measuring the atmospheric residence time of ^{210}Po varies between 15 -75 days with a mean value in the order of 26 □ 3 days. ^{210}Pb is continuously deposited from the atmosphere in association with aerosols at a rate of about 55 $Bq.m^{-2}.a^{-1}$ over Scandinavia (F. Eldaoushy and R. Garciatenorio, 1988). Generally, atmospheric ^{210}Pb concentrations are related to if the underlying surface is terrestrial area, oceanic areas including islands. Permafrost, ice and snow covered surface reduce the atmospheric ^{210}Pb concentrations (F. El-Daoushy, 1988).

Atmospheric fallout of ^{210}Po is normally assumed to be constant at any given site, measured on timescales of a year or more. The ^{210}Po flux may, however, vary spatially by an order of magnitude, depending on factors such as rainfall and geographical location.

These basic concepts have been investigated by carrying out direct measurements of ^{210}Po fallout on both short and long timescales, and by developing mathematical models of ^{210}Po in the atmosphere (G. T. Piliposian and P. G. Appleby, 2003) .

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Direct measurements of ^{210}Po fallout on weekly or monthly timescales using bulk deposition collectors have been made at a number of sites in Europe and beyond. Indirect measurements of the mean atmospheric ^{210}Po flux over several decades have been made using cumulative deposits in selected soil cores. Simplified models of the evolution of the vertical distribution of ^{222}Rn, ^{210}Po and their daughter products ^{210}Bi and ^{210}Po in a vertical column of air moving over the Earth's surface have been developed and used to model geographical variations in the ^{210}Po flux long-range transport is of major importance when modelling atmospheric fallout in regional domains (P. G. Appleby, 2008).

^{210}Po and ^{210}Po in soil

Airborne particles with attached ^{210}Pb and ^{210}Po are carried back to the earth's surface through fallout resulting in the deposition and accumulation of the final long-lived ^{210}Pb (22.3 a) which decay to ^{210}Bi (5 d), ^{210}Po (140d) and finally to stable ^{210}Pb in plants or the top soil.

Soil consists of particles of different minerals as well as organic matter in various stages of degradation. Soil is one of the most important components in the evaluation of radionuclide migration behaviour and distribution of uranium series radionuclides and thus ^{210} Po and ^{210}Pb in a terrestrial ecosystem.

The ^{210}Po in soils may originate either as a product of the radioactive decay of radionuclides of ^{238}U series present in the soil (supported) or the result of the precipitation of radon decay products from the atmosphere (unsupported). The ^{210}Po content of soil varies with soil type. The levels of ^{210}Pb and ^{210}Po contained in the top layer of soil can be correlated with the amount of atmospheric precipitation. In soils, ^{210}Po is in equilibrium with ^{210}Pb, suggesting that the ^{210}Pb in the soil is the main source of ^{210}Po irreversibly adsorbed on clay and organic colloids in the soil (Y. D. Parfenov, 1974).

The vertical distribution of ^{226}Ra and ^{210}Po has been investigated in the cultivated soils of the Buyuk Menderes Basin in Turkey (S. Akyil et al., 2008). The activity concentrations of soil cores range from 80 to 1170 Bq/kg for ^{226}Ra and from 10 to 870 Bq/kg for ^{210}Po with the depth. Analysis of the vertical soil profiles indicate that the activity concentrations of ^{226}Ra and ^{210}Po for soil strata at all the sites was not extremely changed with depth. The activity concentrations of ^{210}Po are slightly high in the surface of all the soil cores while the activity concentrations of ^{226}Ra are slightly low.

The distribution of ^{210}Pb and ^{210}Po concentrations in soil from two districts located at the south-west region of Syria have recently been studied (M. S. Al-Masri et al., 2008). The soil of Dara'a fields was originated by the erosion and degradation of basaltic volcanic primary rocks, which formed dark brown soils, while the soil of Daher Al-Jabal site is clay texture>30% and reddish brown or dark red colour. Due to soil cultivation homogeneous distribution of the studied natural radionuclides with depth has been observed. But differences were found for ^{210}Pb and ^{210}Po concentrations, where high levels of these two radionuclides were observed in the top soil layers. The ^{210}Po concentration varied between 1.2 and 110 Bq/kg. The highest concentration (110 Bq/kg) was also found to be in the grape field soil of Daher Al-Jabal, which has also the highest concentration of ^{226}Ra (36 Bq/kg and ^{238}U (33 Bq/kg) (M. S. Al-Masri et al., 2008).

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^{210}Po and ^{210}Pb in ground water

The activity concentration of ^{210}Pb and ^{210}Po in Finnish dug-well water was 13 and 7 mBq/l respectively. The mean activity concentration of ^{210}Pb and ^{210}Po was highest in drilled water 40 mBq/l and 48 mBq/l respectively. From water works it was only 3 mBq/l for both ^{210}Pb and ^{210}Po (P. Vesterbacka, 2007).

Occurrence and distribution of ^{210}Pb and ^{210}Po in selected California groundwater wells the overall activity of ^{210}Pb ranged from 3.7 mBq/l to 1,5 mBq /l and the ^{210}Po activity ranged from 0.25 mBq /l to 555 mBq /l. The activity concentrations of ^{210}Po were always lower than the lead concentrations. (S. R. Ruberu et al., 2007).

The measurements of radioactivity in groundwater samples from Guarani aquifer in Brazil resulted in averages of 7 mBq/l for ^{210}Pb and 2 mBq/l for ^{210}Po (D. M. Bonotto and T. O. Bueno, 2008).

In December 2001 the EC published a Recommendation (K 2001-4580) concerning radon and radon daughter products which, for ^{210}Pb and ^{210}Po, gives a reference maximum concentration of 200 and 100 mBq/l respectively (G. Wallner et al., 2008)

Monitoring of radioactivity in mineral waters collected in Italy resulted in ^{210}Po levels ranged from <0.04 to 21 mBq/l. with 72.5% of samples presents an activity concentration lower than 1.00 mBq/l. Polonium, in fact, has a very low solubility and it only occasionally appears at elevated concentrations in drinking water (D. Desideri et al., 2007).

^{210}Pb - ^{210}Po in vegetation

Uptake of radionuclides from soil to plant is characterized using a transfer factor (TF); the ratio of radionuclide activity concentration per unit mass concentrations (Bq / kg) of plant (C_{plant}) and soil (C_{soil}) respectively.

$$TF = C_{plant} / C_{soil}$$

The TF for a given type of plant and for a given radionuclide can vary considerably from one site to another, with season and with time after contamination. These variations depend on several factors such as the physical and chemical properties of the soil, environmental conditions, and chemical form of the radionuclide in soil (H. Vandenhove et al., 2009).

Usually a linear relation implies a constant ratio of plant concentration to soil concentration (M. S. Al-Masri et al., 2008). But variations in soil properties such as mineralogical composition, organic matter content, pH and fertility components affect uptake resulting in a non-linear relation (A. MartinezAguirre and M. GarciaLeon, 1995; A. MartinezAguirre et al., 1996; A. MartinezAguirre et al., 1997).

Indeed, it has been shown, for radionuclides from the ^{238}U decay chain, the relationship between the CR and the specific activity in the substrate soil seems to be hyperbolic. From experimental measurements of the transfer factor of the plant *Spartinu densiflora* in the

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Odiel march in Spain the following non-linear relationship was found for ^{210}Po: TF = 2.456 $[C_{soil}]^{-0.663}$ (R. Perianez and A. MartinezAguirre, 1997).

Vegetation is also contaminated by ^{210}Pb and ^{210}Po by direct deposition (Ewers et al., 2003). Most of the natural radioactivity content in fresh outdoor living plants is ^{210}Po as the result of the direct deposition of ^{222}Rn daughters from atmospheric precipitation. Already in the 1960th it was estimated that about 80 % of the radioactive materials in plants is due to ^{210}Po as a result of the direct deposition of ^{222}Rn daughters from atmospheric precipitation (W. V. Mayneord et al., 1960; C. R. Hill, 1960).

Plants, however, get radioactive nuclides both by absorption from the soil (supported Po) and by deposition of radioactive fallout on the plants directly (unsupported Po). Therefore, the ^{210}Po in soil contributes to the uncertainty of using plants with root system as monitors for deposition of ^{210}Po.

Studies on transfer of natural radionuclides from soil to plant have been carried out in different regions in the world (K. Bunzl and M. Trautmannsheimer, 1999; L.W. Ewers et al., 2003; A. C. Paul and K. C. Pillai, 1986; V. A. Pulhani et al., 2005; F. V. Tome et al., 2003; B. L. Tracy et al., 1983)

A systematic study has been carried out on 210Po in soil and vegetation samples in the south western Spain. The vertical profile of ^{210}Po in soil, variation of activity with particle size, activity concentration in vegetation, seasonal variation of activity and dry deposition rate were studied.

Studies have also been made for those plants used for human use and consumption (A. MartinezAguirre and M. GarciaLeon, 1995; A. Martinez-Aguirre and M. Garcia-Leon, 1994; A. MartinezAguirre et al., 1996; A. MartinezAguirre et al., 1997; A. Martinez-Aguirre and R. Perianez, 1998, 1999; S. L. Simon and L. Fraley, 1984; S. L. Simon and S. A. Ibrahim, 1987; L.H. Staven et al., 2003).

^{210}Po and ^{210}Po in tobacco

In 1964 it was pointed out that appreciable concentration of polonium-210 occur in tobacco that is inhaled by smoking (E. P. Radford and V. R. Hunt, 1964). Since then, several investigators have studied both the sources and behaviour of ^{210}Po and ^{210}Pb in relation to smoking, and the biological effects of these on lung tissues and other organs (S. N. A. Tahir and A. S. Alaamer, 2008; T. Kovacs et al., 2007; M. N. Al-Arifi et al., 2006; S. Schick and S. Glantz, 2005; A. E. M. Khater, 2004; A. C. Peres and G. Hiromoto, 2002; B. Skwarzec et al., 2001b; G. Yaprak and B. Uysal, 1998; F. P. Carvalho, 1995; Y. Takizawa et al., 1992; M. Sakanoue and T. Masuda, 1985; E. P. Radford, 1974; R. Soremark and V. R. Hunt, 1967).

High activity concentrations of ^{210}Po and ^{210}Pb are found in tobacco and its products. The results of Tso (1966) indicate that the principal source of ^{210}Pb, and thus of ^{210}Po, in tobacco is the soil and that ^{210}Po and ^{210}Pb are absorbed by the plant roots (T. C. Tso et al., 1966). This finding differs from that of by Berger et al. (1965), but is in agreement with Marsden (1964) that the contribution of ^{210}Po from fallout to the total activity of the plant is minor compared to the ^{210}Po absorbed from the soil by the roots (K. C. Berger et al., 1965; E.

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Marsden, 1964). However, other factors may also contribute to the final concentration of ^{210}Po in tobacco (T. C. Tso et al., 1966).

In Figure 1 is displayed the results of measuring activity concentrations of ^{210}Po and ^{210}Pb in tobacco harvested at different years in various countries USA(T. C. Tso et al., 1964; T. C. Tso et al., 1966); NZ and USA(L. P. Gregory, 1965); Greece (A. Savidou et al., 2006) and Pakistan (S. N. A. Tahir and A. S. Alaamer, 2008).

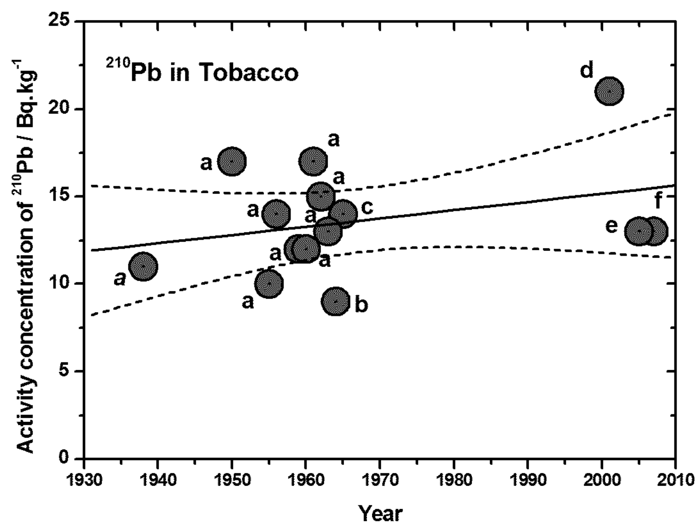

Figure 1.- *Activity concentrations in tobacco harvested at different years in various countries.*
a(USA)(T. C. Tso et al., 1964); b(USA)(T. C. Tso et al., 1966); c(NZ,USA)(L. P. Gregory, 1965); d(A. C. Peres and G. Hiromoto, 2002); e(Greece)(A. Savidou et al., 2006); f(Pakistan) (S. N. A. Tahir and A. S. Alaamer, 2008)

The concentrations of ^{210}Pb and ^{210}Po in the air ways and the lung tissues increase by smoking of tobacco which contributes to an increase in the internal radiation dose. This might influence the incidence of lung cancer observed among smokers, why cigarette packs should carry a radiation-exposure warning label (M. E. Muggli et al., 2008).

The activity concentration of ^{210}Po has been studied in the samples of fresh tobacco, filters before and after, smoke and ash from cigarettes of various brands produced in Poland (B. Skwarzec et al., 2001a). High ^{210}Po activity contents of about 20 mBq per cigarette was found in fresh tobacco of both low-quality brands as well as in the high-quality cigarette Some brand, however, has a ^{210}Po activity content as low as 4 mBq per cigarette. The mean value ^{210}Po activity in the fresh tobacco of 14 different cigarette brands was 13±8 mBq per cigarette. Prior to smoking, the 210Po activity of filters was small, with values in the range of 0.02 - 0.76 mBq . It was observed that, during smoking, the filters absorbed only about 0.1–16.5% (mean value 2.5) of the polonium contained in the tobacco.

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Volatilization of polonium is evidenced by the low ^{210}Po activity measured in cigarette ash as compared with the total content in an unburned cigarette. The fraction of ^{210}Po contents of the ash varied between 4 - 65% of the content in fresh tobacco (average 26 ± 24 %) of different brands.

This is due to the different tobacco-burning temperatures of smoking (between 500 and 700ºC), which cause ^{210}Po to sublime more or less into the smoke which is inhaled by the smoker (Martell, 1975). Therefore the activity of ^{210}Po in smoke inhaled from a cigarette varied a lot, ranged from 2 to 23 mBq with an average of 10 ± 6 mBq . The level from a specific brand should therefore be declared on the cigarette package.

The average (range) activity concentration of Po-210 in cigarette tobacco of Greece was 16.6 (9.7-22.5) mBq/cigarette. The average percentages of Po-210 content in fresh tobacco plus wrapping paper that were recovered by post-smoking filters, ash and smoke were 4.6, 20.7 and 74.7, respectively. It was estimated that cigarette smokers are inhaling on average 6 mBq per cigarette of ^{210}Po and ^{210}Pb each which is in good agreement with the value given above (A. E. M. Khater, 2004).

^{210}Po and ^{210}Pb in mosses

Mosses such as *Polytrichum* and *Sphagnum* have no uptake from soil but are occasionally submerged with surface water. Beard mosses *Alectroria*, however, might be contaminated by re-suspension from soil. A study of various moss samples collected 1979-1980 around Lilljuhatten in Sweden. The min and max level of ^{210}Po in *Polytrichum* was 300-960 Bq/kg dw, in *Sphagnum* 185-700 Bq/kg dw and in Alectoria 570-640 Bq/kg (E. Holm et al., 1981).

Moss samples from the environment of Kaiga nuclear power plant site in the south western region of India were analyzed for ^{210}Po. *Pterobryopsis tumida* a plant of the moss family, shows a very high level of ^{210}Po activity - 2724 +/- 13 Bq /kg dry wt. Seasonal variation studies show elevated levels of ^{210}Po in vegetation during winter. The annual dry deposition rate of Po-210 was 53.4 $Bq.m^{-2}.a^{-1}$. (N. Karunakara et al., 2000)

In Gokova region where Yatagan is located there are three major coal-fired power plants and they cause some pollution in the surroundings. The mosses *Grimmia pulvinata*, *Hypnum cupressiforme* were investigated for potential use as bioindicators for ^{210}Po and ^{210}Pb deposition. The maximum ^{210}Po and ^{210}Pb activities were observed around the hill close to ash stacks. The capture efficiency was the highest in one of the moss species, *G. pulvinata* with the activity concentration ranges of 600 - 1228 and 446 - 650 Bq /kg for ^{210}Po and ^{210}Pb , respectively The corresponding annual Pb-210 flux of 103 $Bq.m^{-2}.a^{-1}$ is high compared to estimates of the atmospheric flux given in literature for the same region. (A. Ugur et al., 2003). The mosses *Grimmia pulvinata, Hypnum cupressiforme* were also analyzed for Pb (A. Ugur et al., 2004) .

Levels of ^{210}Po, ^{210}Pb, and some trace elements such as Pb was studied in two most common mosses (*Lycopodium cernuum* and *Funaria hygrometrica*) distributed in the eastern Mediterranean sea region (Syrian coastal mountains series), The activity concentration of ^{210}Po and ^{210}Pb were found to be 1322 and 1140 Bq/kg dry wt. in *L. cernuum*, respectively. The moss species *F hygrometrica* was found to have even higher values of ^{210}Po and ^{210}Pb

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with activity concentrations of 2392 and 2119 Bq/kg dry wt., respectively. Stable lead concentration in *L. cernuum* was found to be in the range of 5-86.6 mg/kg dry wt., while *F. hygrometrica* samples were found to contain around 58 mg/kg dry wt.. Both plants seem to accumulate lead from the atmosphere where vehicle emissions are the main source of lead in the region (M. S. Al-Masri et al., 2005).

The high concentrating capacity of mosses collected in region of Katirli mountain in north-western Turkey was recently used as bio-indicator of environmental radioactive contamination (A. G. Kahraman et al., 2006).

^{210}Po and ^{210}Pb in peat

Peat mosses are characterized of being primitive plants that grows from the top while the dead bottom develops to peat. Peat is a heterogeneous mixture of partially humified remains of several groups of plants together with inorganic material. The organic material decomposes to insoluble humic acid and lignin derivatives. Humic and fulvic fraction efficiently absorb ^{210}Pb (F. Eldaoushy and R. Garciatenorio, 1988).

Peat cores collected from three sites in the Jura region of Switzerland were analysed for the fallout radionuclides ^{210}Pb. Unsupported ^{210}Pb inventories of the cores were all in the order of 4000 Bq/m^2 and are consistent with the atmospheric deposition about 130 Bq.m^{-2}.a^{-1} (P. G. Appleby et al., 1996).

Data on ^{210}Pb levels in an ombrotrophic peat sequence from a mountain site on the east coast of Ireland are compared with data from a similar sequence at an Atlantic peatland site on the west coast. The unsupported ^{210}Pb inventory at the east coast site was about 6500 Bq/m^2 which is higher than at the west coast 5300 Bq/m^2 and is consistent with the difference in wet deposition at the two sites (D. Gallagher et al., 2000).

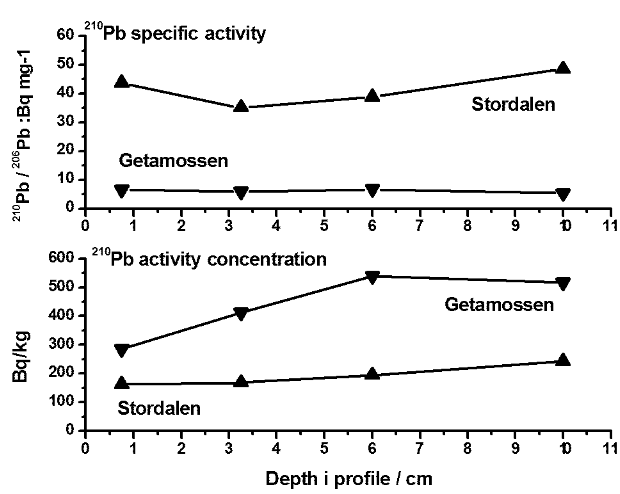

Figure 2.- *The profiles of unsupported ^{210}Pb in peat cores at Stordalen and Getamossen.*

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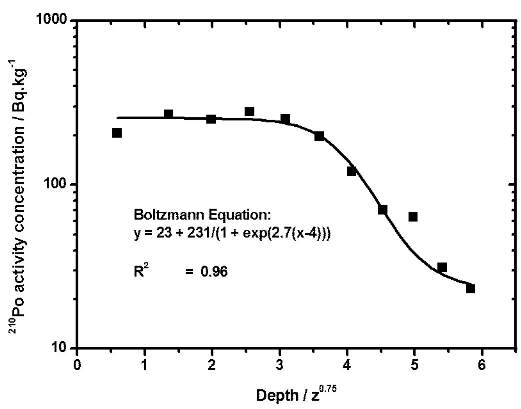
Peat cores of about half a m in depth through bog hummocks with Sphagnum species were sampled in 1979 at Stordalen in the North and Getamossen in the South of Sweden. The unsupported ^{210}Pb inventory at Stordalen was 1300 ± 350 Bq/m^2 and at Getamossen 4000 ± 650 Bq/m^2. The corresponding average activity concentrations in the upper 12 cm of the peat profile were 192±37 Bq/kg and 439 ±117 Bq/kg respectively. The specific activity of ^{210}Pb in the upper 12 cm of the peat profile was 42 ± 8 Bq /mg of stable lead at Stordalen and 6.1± 0.7 Bq /mg of stable lead at Getamossen (N. Malmer and E. Holm, 1984). The corresponding profiles are shown in Figure 2.

Peat cores collected by Holm 2007 in south of Sweden at Toftahult peat land indicate a different depth profile of ^{210}Po and ^{210}Pb as shown in Figures 3 and 4.



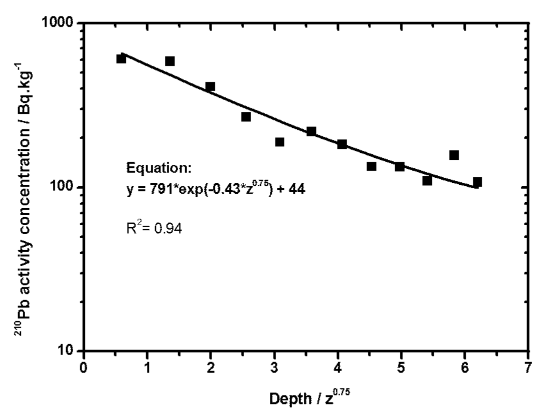


Figure 3.- *^{210}Po activity distribution according to the $z^{3/4}$ depth distribution(B. R. R. Persson et al., 1974)*

Figure 4.- *^{210}Pb activity distribution according to the $z^{3/4}$ depth distribution (B. R. R. Persson et al., 1974)*

^{210}Po and ^{210}Pb in communities of lichen (Cladonia alpestris)

In the studies of the fallout from the atmospheric nuclear weapons tests during 1950th and 60th high activities of ^{137}Cs was found in lichens (G. K. Svensson and K. Lidén, 1965). In the Nordic countries lichens of the Cladonia family are grazed by reindeer which are consumed by man to whom by this way the radioactive fallout was transferred (M. Witkamp, 1966; G. K. Svensson and K. Lidén, 1965). Evidently the same would apply for the natural occurring ^{210}Po and ^{210}Pb (R.B.R. Persson, 1970).

Lichens are slow growing perennials that have high interception potentials for aerosols in precipitation, and therefore contain significantly higher ^{210}Po and ^{210}Pb concentrations than vascular plants (R. B. Holtzman, 1966; P. Kauranen et al., 1971; P. Kauranen and J. K. Miettinen, 1969; Z. Jaworowski, 1969) and fungi (B. Skwarzec and A. Jakusik, 2003). The ^{210}Po /^{210}Pb activity ratio in lichen is typically equal to 1 as ^{210}Po approaches secular equilibrium with ^{210}Pb (L.J.S. Mattsson and R.B.R. Persson, 1971; B. R. Persson, 1970, 1972;

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B. R. R. Persson et al., 1974; P. Kauranen et al., 1971; P. Kauranen and J. K. Miettinen, 1969; P. A. Thomas et al., 1994). In Figure 5 are displayed results from measurement of ^{210}Po in communities of lichen samples from different counties. By assuming radioactive equilibrium the levels are equivalent to ^{210}Pb.

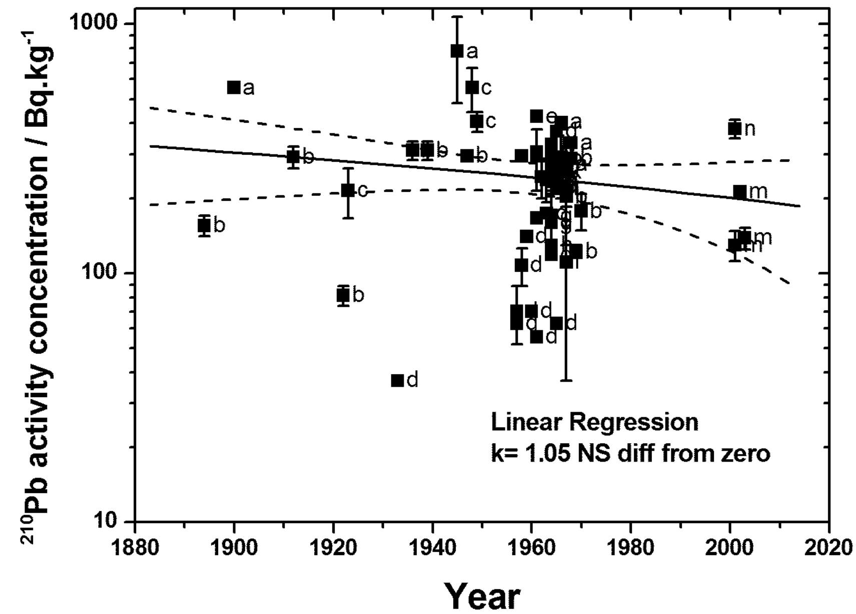

Figure 5.- Results from measurement of ^{210}Po in communities of lichen samples from different counties. By assuming radioactive equilibrium the levels are equivalent to ^{210}Pb.
a)(P. V. Ramzaev et al., 1969); b)(B. R. Persson, 1972); c)(Blanchar.R.l and J. B. Moore, 1970) ; d)(Z. Jaworowski, 1969); e)(R. B. Holtzman, 1966); f)(P. Kauranen and J. K. Miettinen, 1969); g)(Blanchar.Rl and J. B. Moore, 1969); h)(C. R. Hill, 1965); i)(P. Kauranen et al., 1971); j)(R. B. Holtzman, 1966); k)(P. Kauranen et al., 1971); l)(R. B. Holtzman and F. H. Ilcewicz, 1971); m)(L. Skuterud et al., 2005); n)(A. Ugur et al., 2004).

The measurement of ^{210}Po in communities of lichen (*Cladonia alpestris*) at a specific location 63.2□N in central Sweden (Lake Rogen) during 1961-1970 displayed in Figure 6 indicate a more pronounced decrease 3.7% per year although not significant different from zero (p=0.08). In Figure 7 is displayed the specific activity of $^{210}Pb/Pb_{stable}$, which during the same period was quite constant at 23□2 Bq/mg of stable lead. Linear regression wet weights given to the error of each value indicate a quite constant value at 22 Bq/mg during this period (R.B.R. Persson, 1974; B. R. Persson, 1972).

The study stable lead contents in lichens collected since 1882 revealed a steady decrease in the 210Pb/Pb-ratio after 1940. The same indication of an increased world-wide deposition of stable lead since 1940 has also been found by M. Murozumi et al. (1969) and A. Rühling and G. Tyler (1968) in investigations of mosses collected in southern Sweden (M. Murozumi et al., 1969; A. Rühling and G Tyler, 1968). The main source of the increase of the lead content in lichens and mosses seems to be the burning of alkyl lead in automobile fuel (B. R. R. Persson et al., 1974; C. Patterson, 1969)

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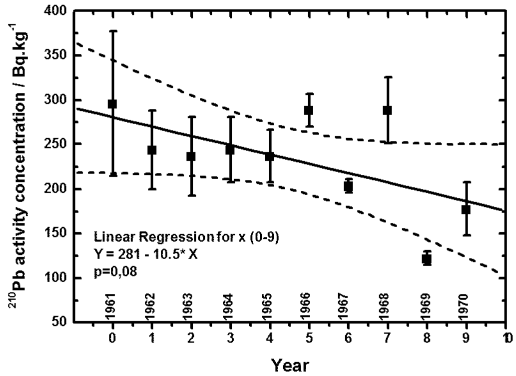

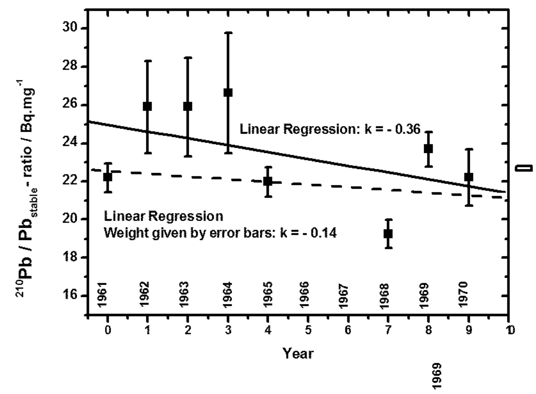

Figure 6.- *Radioactivity concentration of ^{210}Pb in lichen (Cladonia alpestris) collected annually in Sweden 63,2□N.*

Figure 7.- *The ratio of ^{210}Pb-activity concentration to stable lead concentration in lichen (Cladonia alpestris) collected annually in Sweden 63,2□N.*

The variation of the ^{210}Pb/Pb$_{stable}$-ratio with latitude was empirically found to follow an exponential increase with latitude (r =0.86). This might depend on the fact that the fallout of stable lead decreases towards the north of Sweden (G. Tyler, 1970). The ^{210}Pb content of lichens also increases with latitude, but no definite relationship was found in this case (r = 0.39). This might also indicate that the concept of specific activity (^{210}Pb/Pb$_{stable}$-ratio) is less sensitive to sampling variability's and therefore a better indicator of lead pollution-gradients than the stable lead concentration itself.

The value recorded in Sweden in 1970 at a Rogen *63,2□N* agrees very well with the values recorded in Norway in 2001 (L. Skuterud et al., 2005). Thus during the past 40 years there seems to have been a quite constant level of ^{210}Po fallout. The question is, however, if the ^{210}Po fallout will change with future progressed climate changes. A more frequent, regular and careful sampling of lichen, analysis of both ^{210}Po and ^{7}Be and recording of meteorological data (precipitation, temperature etc) might give a possibility to trace trends of climate changes in the terrestrial environment.

Stable lead, ^{210}Pb and ^{210}Po levels were determined in dated sequential growth of thalli of the foliose lichen *Flavoparmelia baltimorensis* from Great Falls and Plummers Island, Maryland. (D. W. Schwartzman et al., 1991) . The average ^{210}Po level derived from their experimental data was estimated to 109±11 Bq/m^2 and the ^{210}Pb level 142±14 Bq/m^2 corresponding to an activity concentration of 64 Bq/kg. The specific activity of lead ^{210}Pb/Pb$_{stable}$ was estimated to 72±11 Bq/mg. Those values are in the same order of magnitude as the other studies referred above. They also confirmed the correlation between the specific activity and the release of stable Pb by using leaded gasoline for automobiles found by (B. R. Persson, 1972; B. R. R. Persson et al., 1974; R.B.R. Persson, 1974) The authors suggest that more rigorous studies of seasonal variations of ^{210}Po/^{210}Pb activity ratio as well as ^{7}Be might be used as an indicator for the effect of climate changes on terrestrial ecological systems particularly in the polar regions (T. Rosswall, 1989, 1994).

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^{210}Po and ^{210}Pb in the food chain lichen-reindeer and man

Various terrestrial food chains contribute to various extent of ^{210}Po intake. Western diet includes from 40 to 400 mBq of polonium-210 per day corresponding to an annual intake of about 10-100 Bq . But Polonium-210 intake can be significantly elevated in some Nordic populations who consume a lot of reindeer.(M. Bolca et al., 2007; K. Bunzl and M. Trautmannsheimer, 1999; P. McDonald et al., 1999; E. E. Santos et al., 2002)

The food-chain: lichen-reindeer man has been used as a model a terrestrial food-chain for human uptake of ^{210}Pb / ^{210}Po and for studying the specific activity ratio of ^{210}Pb/Pb in a historical perspective. This food-chain and other unique ways of polonium-210 transfer to man will be reviewed in a historical perspective.

Already in the 1960th it was observed by several investigators that northern diets, based largely on reindeer or caribou, contain abnormally great amounts of natural "fallout" nuclides ^{210}Po and ^{210}Pb (1-6) (K. Lidén, 1961; P. Kauranen and J. K. Miettinen, 1969; Blanchar.Rl and J. B. Moore, 1970; B. R. R. Persson et al., 1974; M. Baskaran et al., 1991; C. R. Macdonald et al., 1996)

Measurements of ^{210}Po activity concentrations in reindeer meet samples from Finnish Lapland showed activity concentrations of about 3 Bq/kg w.w. in autumn, 5 Bq/kg w.w. in winter and 12 Bq/kg w.w. in spring. For ^{210}Pb the annual average activity concentrations in reindeer meet was ten times lower, 0.22±0.04 Bq/kg ww , with less seasonal fluctuation.
This maintains a high ^{210}Po concentration in soft tissues of reindeer breeding Lapps (about 12 times higher than in southern Finns (P. Kauranen and J. K. Miettinen, 1969). This was shown to be true also for Alaskan residents consuming caribou or reindeer meat (Blanchar.Rl and J. B. Moore, 1970).

^{210}Po average activity concentrations in Swedish reindeer meet samples from animals slaughtered in March 1970 and 1971 was 10.6±0.6 Bq/kg ww. These animals, two years old, had grazed within the same reindeer breeding district where the lichen sampling area is situated (L.J.S. Mattsson and R.B.R. Persson, 1971; B. R. Persson, 1972; R.B.R. Persson, 1970, 1974)

In bone of reindeer from the island Novaya Zemlya in the arctic sea, the concentrations of the natural ^{210}Po and ^{210}Pb in bone of the recent reindeer (570 +/- 190 Bq/kg) is similar to that which was in the teeth of reindeer a hundred years ago (650-750 Bq/kg) and significantly higher than in the recent mainland reindeer from different regions (180-170 Bq/kg) (G. A. Klevezal et al., 2001). The ^{210}Pb in bone of caribou from 1989-74 was around 610 (490-800) Bq/kg (McDonald 1966).

This compares well with the level of 170 +/- 190 Bq/kg wet weight found in Finnish reindeer during 1964-67 (P. Kauranen and J. K. Miettinen, 1969) **.** By letting 14 volunteers consume 2.0 kg of caribou meat containing 9–40 Bq. kg w.w. And collecting urine and faeces the average GI absorption factor was estimated to 56 +/- 4% . This value agree well with the value of 50% recommended that by the ICRP (P. A. Thomas et al., 2001; ICRP, 1994)

A study of concentrations of Po-210 and Pb-210 in Norwegian reindeer during 2000-2003 focused on potential differences in concentrations of these nuclides in reindeer of different

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ages. Concentrations of ^{210}Po and ^{210}Pb in muscle and liver tissues were comparable to those reported for reindeer in other Nordic areas, with no significant difference in ^{210}Po and ^{210}Pb concentrations between adults and calves or between reindeer from the two different study areas. Mean ^{210}Po activity concentrations in muscle tissue, December 2000, were 23.7±3.7 and 35.5±9.2 Bq/ kg dry weight in calves and females (7 years) respectively, not significantly different from those in 2002. These value corresponds to 6 ± 1 and 9.2 ±2.3 Bq/ kg wet weight respectively, which is in agreement with the values recorded in Sweden 1970-71(ICRP, 1994; L.J.S. Mattsson and R.B.R. Persson, 1971; B. R. Persson, 1972; R.B.R. Persson, 1970, 1974)

The ^{210}Pb and ^{210}Po activity concentrations determined in muscle and liver tissue from Norway were similar to values reported from other Nordic areas (B. R. Persson, 1972; R.B.R. Persson, 1974; L. Skuterud et al., 2005; J. P. Gwynn et al., 2006; P. Kauranen and J. K. Miettinen, 1969; B.L. Tracy, 1993; P. Kauranen and J. K. Miettinen, 1967).

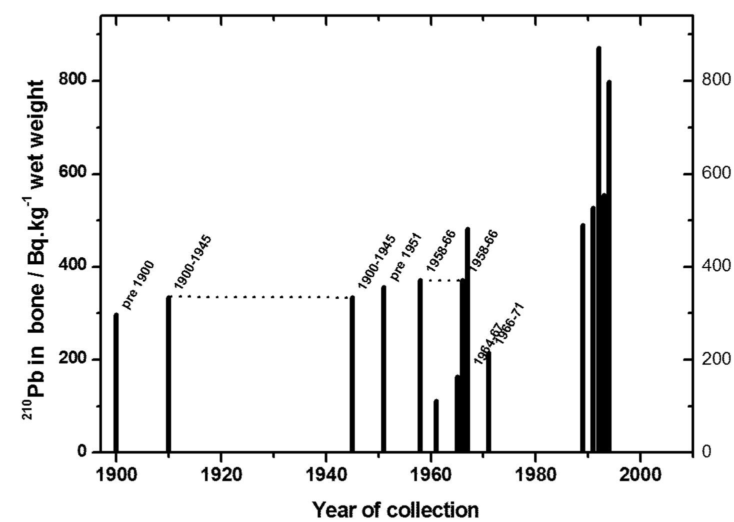

Figure 8.- *^{210}Pb activity concentration in reindeer or caribou bone*

Summary

In Table 1 are compiled a summary of the levels of ^{210}Po and ^{210}Pb activity flux and concentrations in various terrestrial radioecological compartments. The geographical and temporal influence has not been considered due to lack of information. Thus there is a considerable uncertainty in the estimate.

The variations in the analytical method which had been used also influence the precision and accuracy of published data. There seems to be a great need for standardisation of the analytical method. Another wishing is a detailed plan for studying specific annual and seasonal variations at given location in specific compartments over long periods of time. Good example of such a program is the studying of ^{210}Pb fallout in rainwater and sediments in

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UK (P. G. Appleby, 1996, 2008). In order to develop good fallout models, cosmogenic ^{7}Be should also be involved in such a program

^{210}Po and ^{210}Pb activity should be measured both by air filtering and rain water analysis (A. Marenco et al., 1967; J. Paatero and J. Hatakka, 2000). By recording the gamma ray spectrum for analyzing ^{7}Be and gamma emitting radon daughters it might be possible to establish a good model for the relation between the fallout and the local ^{222}Rn flux.

Compartment	**Unit**	**^{210}Pb**	**^{210}Po**	**^{210}Po /^{210}Pb**	**Ref.**
Fallout flux	Bq.m^{-2}.a^{-1}	150 ± 50	100 ± 20	0.21± 0.06	b)
Rain	Bq.l^{-1}	0.1 ± 0.02	0.02 ±0.008	0.30±0.08	c) b)
Surface sediment	Bq.kg^{-1}	600 ± 300			a)
Soil (Turkey)	Bq.kg^{-1}	50 ± 20			d)
Soil (worldwide)	Bq.kg^{-1}	130 ±100			a)
Tobacco	Bq.kg^{-1}	13 ± 2			a)
Vegetables	Bq.kg^{-1}	3 ± 1			e)
Vegetables	Bq.kg^{-1}	0.08	0.1		f)
Milk	Bq.l^{-1}	0.015			f)
Meat	Bq.kg^{-1}	0.08			f)
Drinking Water	Bq.l^{-1}	0.003			f)
Ground and well water	Bq.l^{-1}		0.002	0.3 ± 0.05	b)
River and lakes	Bq.l^{-1}		0.0015	0.45 ± 0.05	b)
Mosses	Bq.kg^{-1}(d.w.)		560 ± 280		a)
Peat 12 cm surface	Bq.kg^{-1}(d.w.)		320 ±170		a)
Lichen	Bq.kg^{-1}(d.w.)		240 ±135		a)
Reindeer meat	Bq.kg^{1}(w.w.)		3.1 ± 1.4		a)
Man:					
Lung	Bq.kg^{-1}(w.w.)		0.200		f)
Liver and Kidney	Bq.kg^{-1}(w.w.)		0.600		f)
Muscle	Bq.kg^{-1}(w.w.)		0.1		f)
Bone	Bq.kg^{-1}(w.w.)		2.4		f)
Annual intake	Bq.a^{-1}	0.69	1.2		f)
Com. Eff. Dose	□Sv. a^{-1}	28	85		f

Table 1.- *Estimates of ^{210}Po and ^{210}Pb activity concentrations in various compartment of the terrestrial environment. The concentrations in tissue of man are given as information although they depend not only from intake of terrestrial origin.*
a) This work; b) (Y. D. Parfenov, 1974); c)(P. G. Appleby, 2008; S. Akyil et al., 2008); d) (S. Akyil et al., 2008); e) (K. Bunzl and M. Trautmannsheimer, 1999); f) (UNSCEAR, 2000);

Unsupported ^{210}Pb inventories of the cores peat collected at various sites were all consistent with the atmospheric deposition about at their location (P. G. Appleby et al., 1996; P. G.

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Appleby, 2008). Thus a program of collecting and analyzing peat core samples at various locations might give a good picture of the geographical and temporal variations. The effect of climate changes might be studied in peat cores by seasonal and annual monitoring over time in addition to fallout measurements.

The level of ^{210}Po and ^{210}Pb activity in tobacco seems so be rather constant over time and geographical distribution with an over all average of 13 ± 2 Bq.kg^{-1}. Due to the impact of ^{210}Po and ^{210}Pb activity levels in tobacco on human health, it should be required to specify the amount of inhaled ^{210}Pb on the cigarette package of each brand (S. N. A. Tahir and A. S. Alaamer, 2008). This might conduce to minimize this level by studying the growing conditions and fallout levels in the tobacco fields (M. E. Muggli et al., 2008).

The food chain lichen-reindeer and man is a unique model for studying the uptake and retention in man. As seen in Table 1 the level of ^{210}Po and ^{210}Pb activity in lichen is quite high and the level in reindeer meat is much higher than in ordinary flesh and meat on the market. Although reindeer meat is regularly consumed by a minor part of the population more efforts should be taken to support studying this food chain (L. Skuterud et al., 2005; P. A. Thomas et al., 2001; G. A. Klevezal et al., 2001) .

The level of ^{210}Po and ^{210}Pb activity in drinking water and in most common food items seems to be low. Fresh vegetables exposed to fallout might be important to monitor more carefully (M. Bolca et al., 2007; E. E. Santos et al., 2002; P. McDonald et al., 1999; P. L. Santos et al., 1993; K. Bunzl and M. Trautmannsheimer, 1999).

In conclusion the ^{210}Po and ^{210}Pb activity in terrestrial radioecological system has great potential for future research in monitoring the effects of climate changes and to improve human health by reducing the levels in tobacco.

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